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Tampa Bay Area Photochemical Oxidant Study. Appendix C. Isoprene Emission Rates from Live Oak

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TAMPA BAY AREA
PHOTOCHEMICAL OXIDANT STUDY

APRIL 1978

FINAL REPORT

APPENDIX C

ISOPRENE EMISSION RATES
FROM LIVE OAK

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ATLANTA, GEORGIA 30308

ISOPRENE EMISSION RATES FROM LIVE OAK

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ABSTRACT

There is growing awareness about the role of vegetation as a source of reactive hydrocarbons that may serve as photochemical oxidant precursors. A study was designed to assess the influence, independently, of variable light and temperature on isoprene emissions from live oak (Quercus virginiana, Mill.). Plants were conditioned in a growth chamber, then transferred to an environmentally controlled gas-exchange chamber. Samples of the chamber atmosphere were collected, isoprene was concentrated cryogenically and measured by gas chromatography. The logistic function was used to model isoprene emission rates. Under regimes of low temperature (20°C) or darkness, isoprene emissions were lowest. With increasing light intensity or temperature the concentration of isoprene increased, reaching maxima at 800 μEinsteins/m^2/sec and 40-44°C, respectively. Higher temperatures caused a large decrease in emissions. Since the emission of isoprene is light saturated at moderate intensities, temperature appears to be the main factor controlling emissions during most of the day.

INTRODUCTION

High levels of ozone have been measured in urban, rural and remote locations well away from significant anthropogenic sources of oxidant precurors. These elevated levels in rural and remote areas could result from transport into these areas and/or the photooxidation of biogenic hydrocarbons. It is unclear which process is more important, in part, because biogenic emission rates of reactive hydrocarbons are not adequately known. Isoprene (2-methyl-1,3-butadiene) has been identified as a volatile emission product from at least 35 plant species representing 16 families (Table 1). The biogenic production of isoprene is light dependent (Rasmussen and Jones 1973; Sanadze and Kalandadze, 1966b; Sanadze and Kursanov, 1966) and increases with light intensity until the process controlling emissions are light saturated (Sanadze and Kalandadze, 1966a). The action spectra of isoprene production exhibits a maximum in the red wavelengths (Rasmussen and Jones, 1973; Sandze and Kalandadze, 1966b). The temperature coefficient (Q_{10}) of isoprene production varies between 2.8 and 3.6; emissions increase with temperature reaching a maximum between 35 and 43°C and decreases sharply at temperatures above 40 to 43°C (Sanadze and Kalandadze, 1966a; Rasmussen and Jones, 1973).

Previous studies (i.e. Sanadze and Kalandadze, 1966a and 1966b; Rasmussen and Jones, 1973) used detached leaves or leaf discs. The objectives of this study were, (1) to determine isoprene emission rates from intact plants under

carefully controlled environmental conditions, (2) to determine the independent influence of light and temperature on emission rates, and (3) to estimate algorithms for isoprene emissions which could be used to standardize emission rates determined in the field.

METHODS AND MATERIALS

PLANT CULTURE. Live oak (Quercus virginiana, Mill.) plants were obtained from the Division of Forestry, Florida Department of Agriculture and Consumer Services as bare root seedlings and potted in 15-cm pots in a Jiffy-mix; perlite* (1:2;V:V) mixture. After initial leaf drop and subsequent appearance of new growth, the oak plants were cultured in a greenhouse at maximum day/night temperatures of 23° and 18°C, respectively. Sunlight was supplemented and the photoperiod extended to 16 hr per day with light from HID sodium vapor lamps. The plants received 1/2 strength modified Hoaglund's nutrient solution daily. At least 4 weeks before sampling, oak plants were placed in a growth chamber and conditioned at maximum day/night temperatures of 27° and 18°C, respectively, with a 16 hr light period. When samples were taken, the plants had both mature and young expanding leaves.

GAS-EXCHANGE SYSTEM. The gas-exchange system used to determine emission rates (Figure 1) consisted of a 1) gas-exchange chamber that enclosed the foliage, 2) an air flow system that controlled CO_2 concentration, dew point and provided hydrocarbon-free air (pure air source) to the gas-exchange

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chamber, and 3) a monitoring system that measured CO_2 concentration and dewpoint of the atmosphere entering and exiting the plant chamber, light intensity within the plant chamber, and air and leaf temperature.

GAS-EXCHANGE CHAMBER. The gas-exchange chamber was similar to one described by Huang, et al. (1975) with the mixing characteristics of a constant-stirred-tank reactor (Rogers, et al., 1977). The gas-exchange chamber (Figure 2) was designed for use in a controlled environment chamber which regulated light and cooling. In each experiment the upper chamber was removed to insert the pot-root mass into the lower chamber. The two halves of the 24 cm diam disc with center cut out were fitted around the stem, set in place, and sealed to the stem and chamber with modeling clay. The upper chamber was placed over the plant without injuring leaves and sealed to the chamber base. Both upper and lower chambers contained impellers with exterior motors and the upper chamber contained 2.5 cm high baffles arranged equidistant around the walls of the chamber to insure well mixed air. Heating element and temperature sensor were connected to a Love model 48* temperature controller. Within the upper chamber air temperature was monitored with a shielded thermocouple, leaf temperature with a thermistor clipped to the undersurface of the leaf and light (400 - 700 nm) with a Lambda Instruments model LI-190 SR Quantum Sensor.* Various light levels were obtained by a stepwise increase in the number of incandescent and fluorescent lights in the controlled environment chamber and the addition of a sodium vapor lamp.

AIR FLOW SYSTEM. Air was pumped through an Aadco* pure air generator to remove hydrocarbons, CO_2 and reduce the dew point (Figure 1). Carbon dioxide and water vapor were then added back to the air stream to obtain desired levels. The dew point of the air entering the gas-exchange chamber was held constant during each experimental run. Inlet dew points ranged between -2 and 6°C and were obtained by mixing dry air from the pure air generator with humidified air. The dewpoint of air exiting the chamber varied between 15 and 32°C depending on experimental conditions. Air flow into the inlet port of the gas-exchange chamber was adjusted by a valve, monitored by a flowmeter and ranged from 2 to 5 l/min depending on plant size and environmental conditions. Air samples for CO_2 dew point and hydrocarbon analysis were taken from sample ports at the chamber's inlet and outlet, respectively. Carbon dioxide concentration and dew point of the air stream were monitored with a infrared gas analyzer and dew point hygrometer, respectively. Inlet and outlet CO_2 concentrations of the air stream ranged from 400 to 600 and 310 to 390 µl/l, respectively. Changes in CO_2 concentrations in the chamber were not large enough to have any significant effect on isoprene emission (Jones and Rasmussen, 1975).

HYDROCARBON SAMPLING AND ANALYSIS. Isoprene was separated using a 3 m x 3.2 mm OD stainless steel column of Porasil E(80/100 mesh)/Alltech CS-10 (10% stationary phase). Analyses were performed isothermally at 90°C using helium (20 cm^3/min) as a carrier gas and the isoprene measured with a flame ionization detector (FID). Since a FID responds linearly to the mass of organic carbon (David, 1974), a 1.01 µl/l isooctane standard was used to calculate the mass of organic carbon emitted as isoprene. Three to six 1 ml isooctane

standards were taken each day with a reproducibility of ± 2%. A 99.9% pure standard of isoprene was used to establish the isoprene retention time and profile.

Twenty ml air samples were collected from the sample ports of the gas-exchange chamber using a 100 ml Pressure LOC* syringe. The samples were injected through a 6 port valve onto a stainless steel trap (61 cm x 0.25 mm ID) immersed in liquid oxygen to concentrate the hydrocarbon sample (Rasmussen, et al., 1974). Following sample injection the stainless steel trap remained in the cryogen for a 4 min period with a helium purge flow (13 cm^3/min) through it. The concentrated sample was volatilized onto the column by rapidly heating the trap in boiling water.

Positive identification of isoprene emissions from live oak was made by a combination of gas chromatography and mass spectrometry. The isoprene mass spectra were compared to the EPA Mass Spectral Search System (MSSS), Registry of Mass Spectral Data (Stenhagen, et al., 1974) and the 99.9% isoprene standard to confirm identification.

EXPERIMENTAL DESIGN. The influence of temperature on isoprene emissions at various light levels was studied by increasing leaf temperature from 20 to 47°C in 4 to 6°C increments at each of 4 light levels (approximately 100, 200, 400, or 800 μEinsteins/m^2/sec). To measure the effect of light intensity on isoprene emissions, light intensity was increased stepwise 0, 100, 200, 400 and 800 μEinsteins/m^2/sec at each of 4 leaf temperatures (29, 35, 40, or 47°C). After each change of light or temperature a 60 min equilibration time was observed before collecting 3 air samples for hydrocarbon analysis. A

minimum of 3 plants was used to develop each temperature or light response curve. After each experiment, leaves were removed from the live oak and dry weight measured after the leaves dried at 70°C for 72 hr.

Isoprene emission rate (μg C/ g dry wt/hr) from the oak was calculated using the following equation:

$$\text{Isoprene emission rate} = \frac{J\Delta Conc}{W}$$

J = air flow rate through the gas-exchange chamber (l/hr)

ΔConc = change in isoprene concentration of air as a result of passage through the gas-exchange chamber (μg/l). There was no isoprene in the air entering the chamber.

W = total leaf dry weight of the plant (g)

DATA ANALYSIS. The relationship between means and standard deviations of samples taken at each light and/or temperature point indicated that the isoprene emission rate was distributed log-normally. Therefore the emission data were transformed to their respective natural logarithms for all statistical analysis. Means of the 3 samples collected at each light and temperature combination were computed and used to estimate the isoprene response curves.

Experimentation (Sanadze and Kalanadze, 1966a; Rasmussen and Jones, 1973) and biological theory suggested that the isoprene response curves should approach asymptotically their maximum values. A 3rd order polynomial, rational and logistic functions were considered as models for the response curves. The logistic function was selected because it had good asymptotic properties and best fit the data.

The equation for the 4 parameter logistic function is:

$$F(x) = \frac{A}{1 + \exp[-B(x-C)]} + D$$

A = the difference between the minimum and maximum

B = shape parameter which determines how quickly the function goes from its minimum to maximum

C = location parameter which determines the point along the X axis where the curve is centered

D = minimum value of function

Since the logistic function is nonlinear in its parameters, a non linear least squares procedure was used to estimate the parameters for the response curves. The algorithm used was Marquardt's (1970) as implemented in a subroutine written by Dr. Larry Male at EPA-CERL Corvallis. The choice of initial parameter estimates, critical in nonlinear estimation, was made by trying various parameter values until plots showed that the resulting curves fit the data reasonably well. Means of isoprene emissions above 44°C were not used to estimate temperature response curves because isoprene production dropped off rapidly at higher temperatures and no longer followed the logistic curve.

RESULTS

Increasing the light at all temperatures increased significantly isoprene emission rates until maxima were reached (Figure 3). The parameter estimates for the logistic curves illustrated in Figure 3 are listed in Table 2. In the dark at all temperatures isoprene emissions were low (0.5 μg C/g dry wt/hr or less). As light intensity increased isoprene emission increased significantly to about 75 μg C/g dry wt/hr at 40°C and 800 μEinsteins/m^2/sec. However, at 47°C increasing light elicited only a slight increase in isoprene emission (2 μg C/g dry wt/hr). As the light intensity increased, light saturation of isoprene emission process was approached (within 5%) or reached. Light intensities above the saturating light level would not significantly increase isoprene emission rates.

Increasing temperature increased isoprene emission rates at the four light levels (Figure 4). The parameter estimates for the logistic curves illustrated in Figure 4 are listed in Table 3. Isoprene emissions increased with temperature at each light level but the increases were greater at the higher light intensities. Maximum isoprene emissions occurred between 40 and 44°C with the largest emission (about 120 μg C/g dry wt/hr) at the high light level and 44°C. At higher temperatures (above 44°C) for all light levels there was a sharp decline in isoprene emission to 2 μg C/g dry wt/hr or less. When the higher temperatures persisted for several hours, marginal leaf

necrosis developed. No emission data were collected below 27°C at the high light intensity because the gas-exchange chamber could not be cooled to lower temperatures under this radiant-energy load.

To determine if isoprene emission rates varied significantly over time for plants held under constant environmental conditions, emission rates were measured over a 5 to 6 hr period (Table 4). The flux estimates varied ± 4 and ± 2 μg C/g dry wt/hr about the means for plants 1 and 2, respectively. These variations in emission rates are essentially insignificant compared to the changes caused by altering the environment.

To illustrate the magnitude of plant-to-plant variability, emission rates were measured for 18 different plants under 3 different light levels at a leaf temperature of 35°C (Table 5). At each light intensity most emission rate estimates were close to the mean; only a few plants deviated substantially. Based on this limited sampling, plant variability appeared to range between 20 and 45%.

Despite plant-to-plant variability, the logistic response curves fit the data closely (Figures 5 and 6) and this is supported by the multiple correlation coefficients (R^2) listed in Tables 2 and 3 which are generally 0.85 or greater. Each graph illustrates the logistic response curve and the means used to estimate that curve. Comparisons of the estimated response curves to the data illustrate the generally good agreement. Only at 47° (Figure 5) was there a poor fit of the data to the response curve. This occurred under conditions of very low emissions and unusually high temperatures.

DISCUSSION

The light dependency of isoprene emissions and the light saturation response of live oak were similar to other plant species (Sanadze and Kalandadze, 1966a; Rasmussen and Jones, 1973). Because the isoprene emission process was light saturated at relatively low light intensities (approximately 800 μEinstiens/m^2/sec) many leaves of a canopy would be light saturated for most of the day. The primary importance of light to isoprene emission is to provide the photochemical energy to drive the biosynthetic pathways of isoprene formation. The small amounts of isoprene emitted during dark hours have been previously reported (Rasmussen and Jones, 1973). It is not known if these emissions result from isoprene produced in the dark or if they are residual from the preceding day.

The temperature influence of isoprene emissions from live oak was similar to responses reported for other species (Sanadze and Kalandadze, 1966a; Rasmussen and Jones, 1973). Leaves which are strongly sunlit can be considerably warmer than air temperature, while leaves within the canopy may be cooler than air temperature. The high leaf temperatures (greater than 44°C) associated with decreased emissions could occur under field conditions on leaves in full sun. The high temperature (above 44°C) decrease in isoprene emissions may be associated with the temperature inactivation of the metabolic processes that form isoprene.

To model typical diurnal isoprene emission patterns the environmental conditions for an average of summer days in Tampa, Florida and the estimated isoprene emission rates interpolated from the light intensity and temperature response curves (Figures 3 and 4) were used. Climatic summaries (e.g. Visher, 1954; NOAA, 1974) indicated an average daily solar radiation of approximately 500 Langleys and average maximum and minimum air temperatures of 32 and 20°C, respectively, during the summer. The hourly values for the solar radiation flux were estimated by assuming that the flux varied sinusoidally with a maximum (1.1 Langley/min) at local noon and zero at 0600 and 1800 hr. The average hourly air temperature was inferred from the observed average maximum and minimum temperatures and what is known about typical diurnal temperature cycles. Empirical results in our laboratory indicated that the temperature of sunlit leaves was related to the air temperature by $T_l = 1.25\ (T_a + 0.9)$ where T_l is leaf temperature and T_a is air temperature (both in °C). The leaf temperature for shaded leaves was assumed to be 1°C less than air temperature.

Estimated hourly emission rates of isoprene for oak trees in the vicinity of Tampa, Florida are shown in Figure 7. As expected, the predicted maximum emission rates occurred, during midday. During early morning and late afternoon hours when temperatures are moderate and leaves are not light saturated, light is the main factor controlling emission rates. However, during most of the day, the leaves of the outer canopy are light saturated and varying air temperature controls emission rates. In contrast, for heavily shaded leaves (not light saturated) the light intensity is important throughout the day. More than 80% of the daily isoprene emissions from oak trees is expected

to occur after 1000 hours. The isoprene emissions estimated from the model and reported in Figures 3, 4, 5 and 6 were similar to emission rates measured on oak in the Tampa, Florida, area (P.R. Zimmerman, personal communication). The estimated emission rates in Figure 7 could be altered by assuming different environmental conditions at the leaf or by changing the proportions of leaves that are sunlight to shaded leaves.

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TABLE I: PLANT SPECIES IN WHICH ISOPRENE HAS BEEN REPORTED

SCIENTIFIC NAME	COMMON NAME	REFERENCE
Robinia pseudoacacia L.	Black Locust	Sanadze, 1961
Cladrastis lutea (Michx) Koch	Yellow wood	Rasmussen, 1972
Amorpha Pruticosa L.	False indigo	Sanadze, 1969
Inga spectabilis (Vahl) Willd.	-------	Rasmussen, 1970
Rhamnus sp.	Buckthorn	Rasmussen, 1972
Morus sp.	Paper Mulberry	Rasmussen, 1972
Ficus costaricana (Liebn) Mig.	Higo Fig	Rasmussen, 1970
Plantanus occidentalis L.	Sycamore	Rasmussen, 1972
Populus balsamifera L.	Balsam poplar	Rasmussen, 1972
Populus deltoides Bartr.	Eastern Cottonwood	Rasmussen, 1970
Populus nigra L.	Black Poplar	Sanadze and Kalandadze, 1966a
Populus Simonii Carr.	Simon Aspen	Sanadze, 1961
Populus sp.	Aspen	Rasmussen, 1970
Salix alba L.	White Willow	Sanadze, 1961
Salix babylonica	Weeping Willow	Rasmussen, 1970
Quercus borealis Michx.	Northern Red Oak	Jones and Rasmussen, 1975
Quercus iberica Bieb.	Iberian oak	Danadze, and Dilidze, 1961
Quercus rubra L.	Red Oak	Rasmussen, 1970
Hamamelis virginiana L.	Witchhazel	Jones and Rasmussen, 1975
Liquidambar styraciflua, L.	Sweetgum	Rasmussen, 1970
Picea engelmannii Parry	Engelmann Spruce	Rasmussen, 1972
Picea glauca (Moench) Voss	White spruce	Rasmussen, 1970
Picea mariana (Mill) B.S.P.	Black spruce	Rasmussen, 1970
Picea pungens Engl.	Blue Spruce	Rasmussen, 1970
Picea sitchensis (Bong.) Carr.	Sitka Spruce	Rasmussen, 1972
Mangifera indica L.	Mango	Rasmussen, 1970
Buxus sempervirens L.	Common Boxwood	Sanadze, 1969
Eucalyptus camaldulensis L.	Australian river red gum	Rasmussen, 1970
Eucalptus Gunnii Montana Hook.	Cider gum	Rasmussen, and Jones, 1973
Carludovica insignis Duch.	Panama hat palm	Rasmussen, 1970
Bambusa sp.	Bamboo	Rasmussen, 1970
Cocos nicifera L.	Coconut palm	Rasmussen, 1970
Mahonia aquifolia (Pursh) Nutt.	Oregon grape	Rasmussen, 1972
Liriodendron tulipifera L.	Yellow Poplar	Rasmussen, 1972

TABLE 2. PARAMETER ESTIMATES FOR THE LOGISTIC CURVES DETERMINED WITH VARYING LIGHT INTENSITIES*.

Leaf Temperature °C	Parameter Estimates				
	A	B	C	D	R^2
29	8.63	0.00691	54.26	- 4.75	0.91
35	19.12	0.00838	-108.55	-15.12	0.96
40	21.38	0.00570	-207.44	-17.03	0.92
47	9.62	0.00969	-178.08	- 8.93	0.23

* $$\text{Ln (Isoprene)} = \frac{A}{1 + \exp [-B (x-C)]} + D$$

x = Light Intensity $\mu E/m^2/Sec$

TABLE 3. PARAMETER ESTIMATES FOR THE LOGISTIC CURVES DETERMINED WITH VARYING TEMPERATURES*.

Light Intensity $\mu E/M^2/sec$	Parameter Estimates				
	A	B	C	D	R^2
100	1.55	0.20	25.17	0.16	0.85
200	1.58	0.79	32.97	1.60	0.90
400	1.87	0.35	30.27	1.86	0.88
800	4.88	0.18	25.26	0.11	0.68

* $$\text{Ln (Isoprene)} = \frac{A}{1 + \exp [-B (x - C)]} + D$$

x = leaf Temperature °C

TABLE 4. ISOPRENE PRODUCTION (μg C/g dry w/hr) UNDER CONSTANT ENVIRONMENTAL CONDITIONS *

Hours+		Plant 1	Plant 2
3		24.5	–
4		32.9	32.9
5		26.4	34.4
6		26.2	34.7
7		27.1	32.5
8		29.5	34.1
9		31.0	35.9
	$\bar{X}$	28.2	34.1

* The plants were held at constant environmental conditions (leaf temperature, 35°C; light intensity, 400 $\mu E/m^2/sec$) and triplicate samples were collected hourly and mean emission rates determined.

\+ Hours after the lights were turned on in the controlled environment chamber.

TABLE 5. COMPARISONS OF ISOPRENE EMISSIONS (μg C/g dry wt/hr) FROM PLANTS AT SEVERAL LIGHT INTENSITIES*

	Light Intensity ($\mu E/m^2/sec$)		
	~200	~400	~800
	18.8	51.8	46.4
	16.6	31.8	108.9
	7.1	33.7	67.4
	24.1	39.3	73.7
	19.2	31.6	60.3
	24.3	44.1	64.1
$\bar{X}$	18.4	38.7	70.1

* Leaf temperature ranged between 33 and 37°C, triplicate samples were collected from each plant and a emission rate determined. Each emission rate estimate was from a different plant.

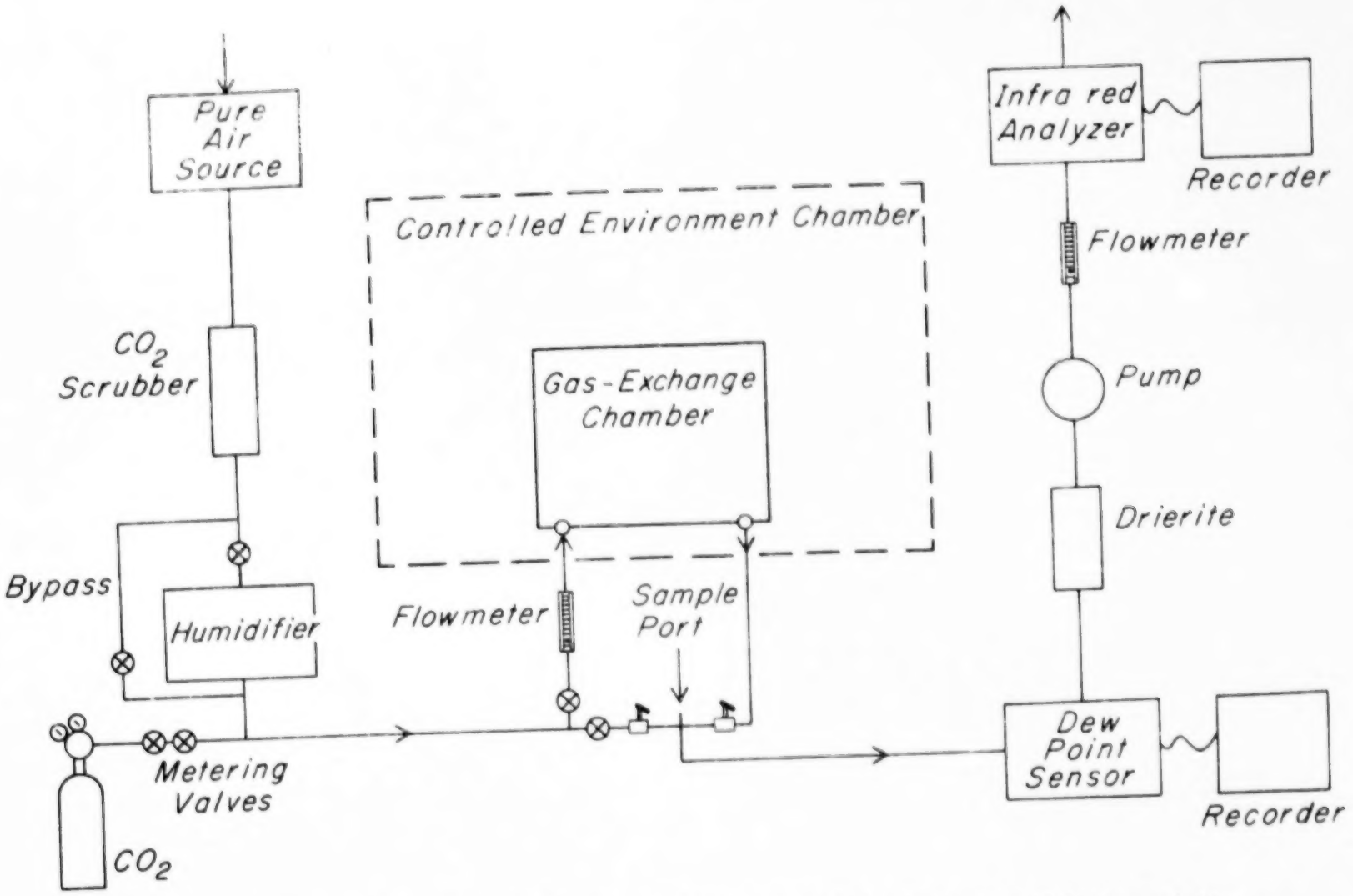

Figure 1. Air flow pattern through the gas exchange chamber and arrangement of instrumentation.

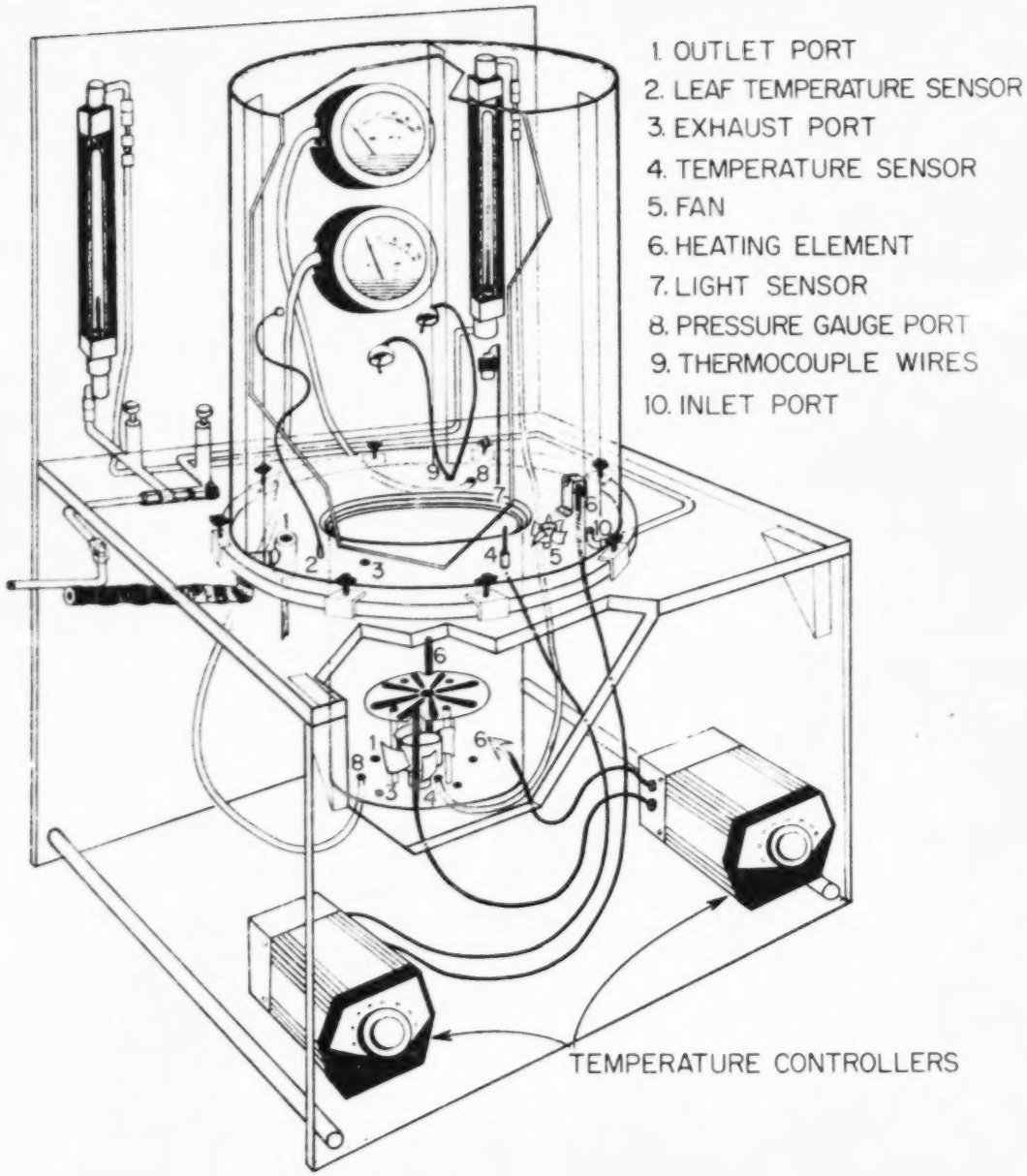

Figure 2. Plant gas-exchange chamber with the major components illustrated. The volumes of the upper and lower chambers were 44.2 and 11.3 liters, respectively.

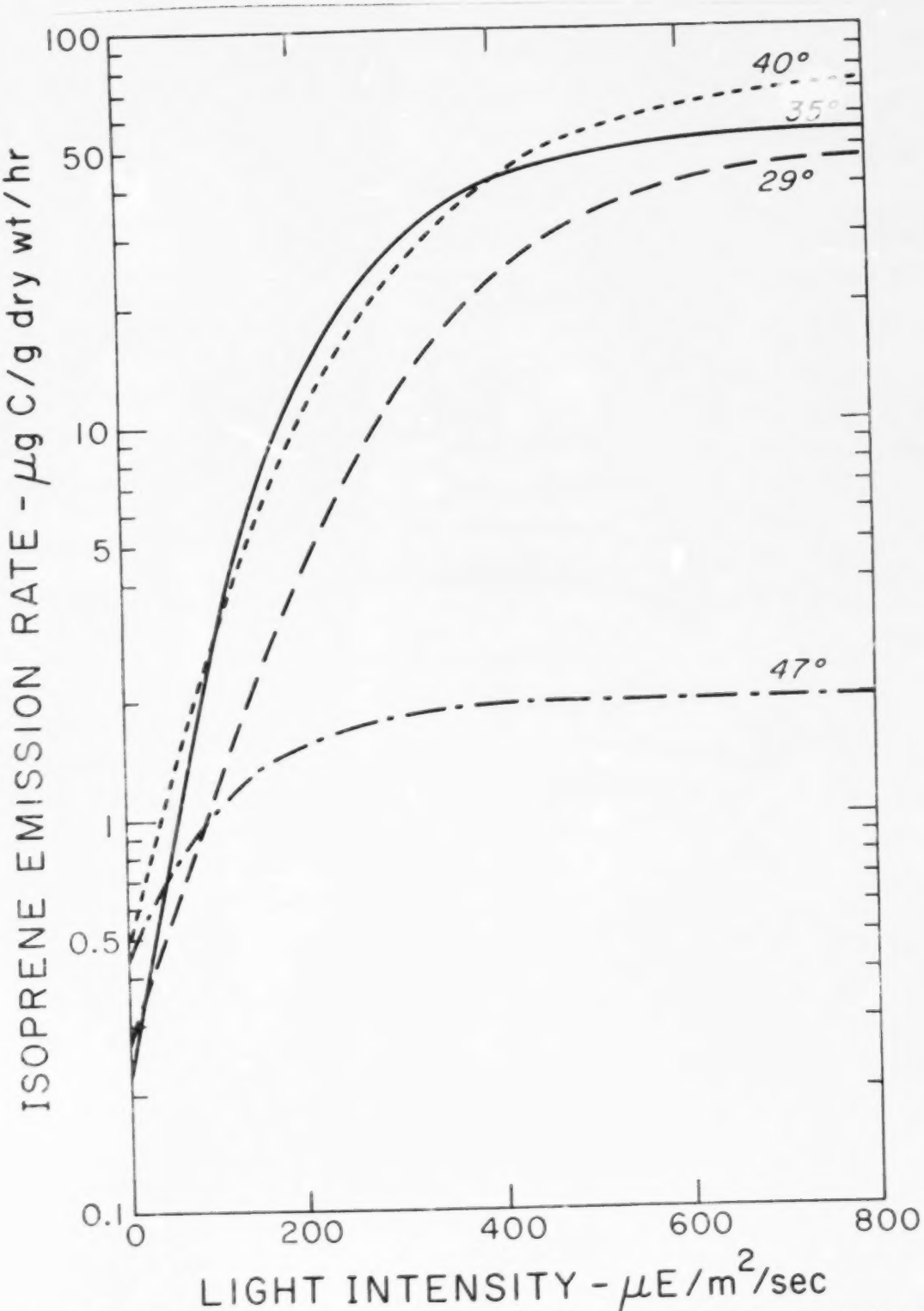

Figure 3. The influence of varying light intensity on isoprene emission rate at various leaf temperatures. The figure is a consolidation of the curves in Figure 5.

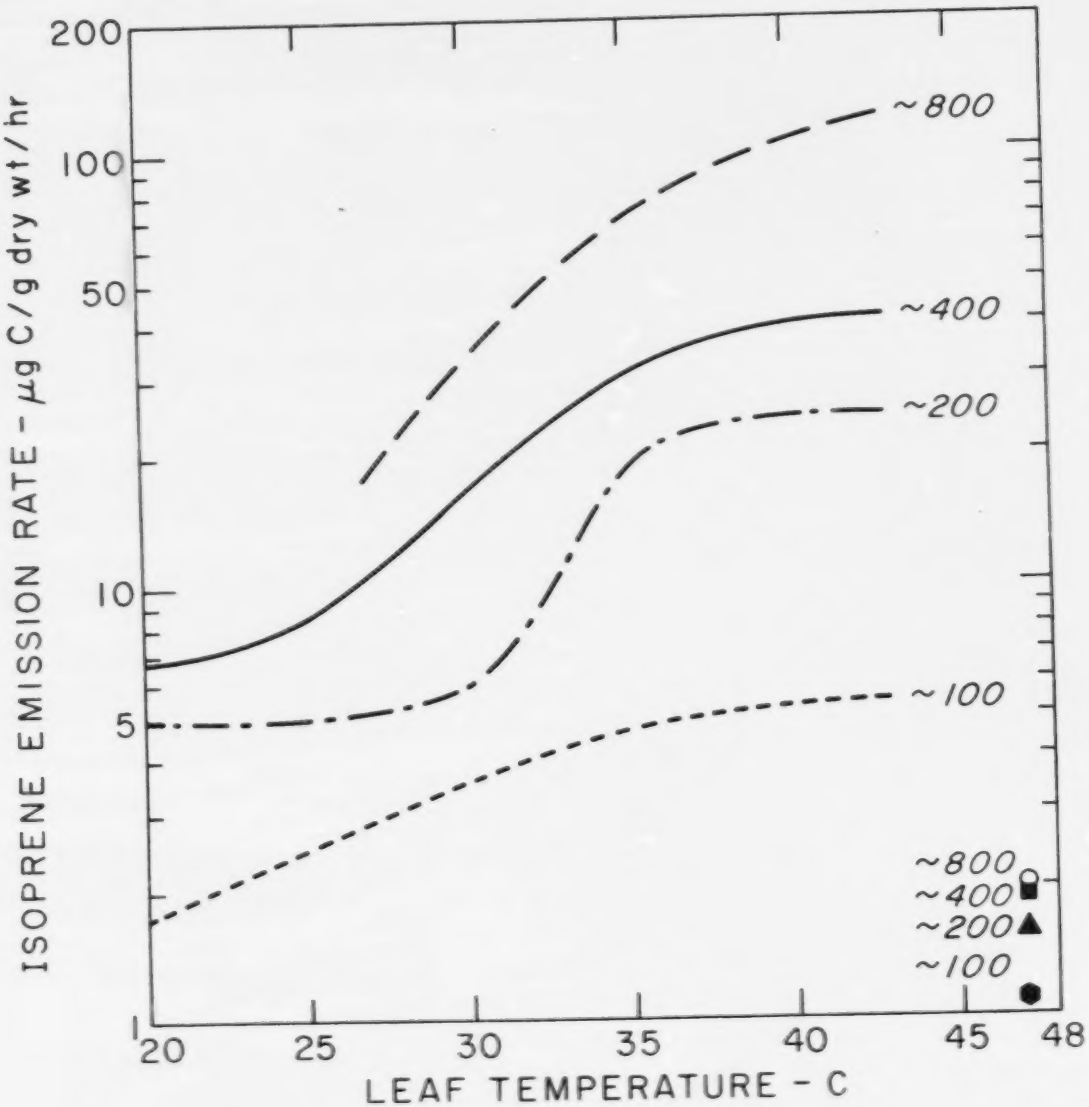

Figure 4. The influence of varying temperature on isoprene emission rate at various light levels. The figure is a consolidation of the curves in Figure 6.

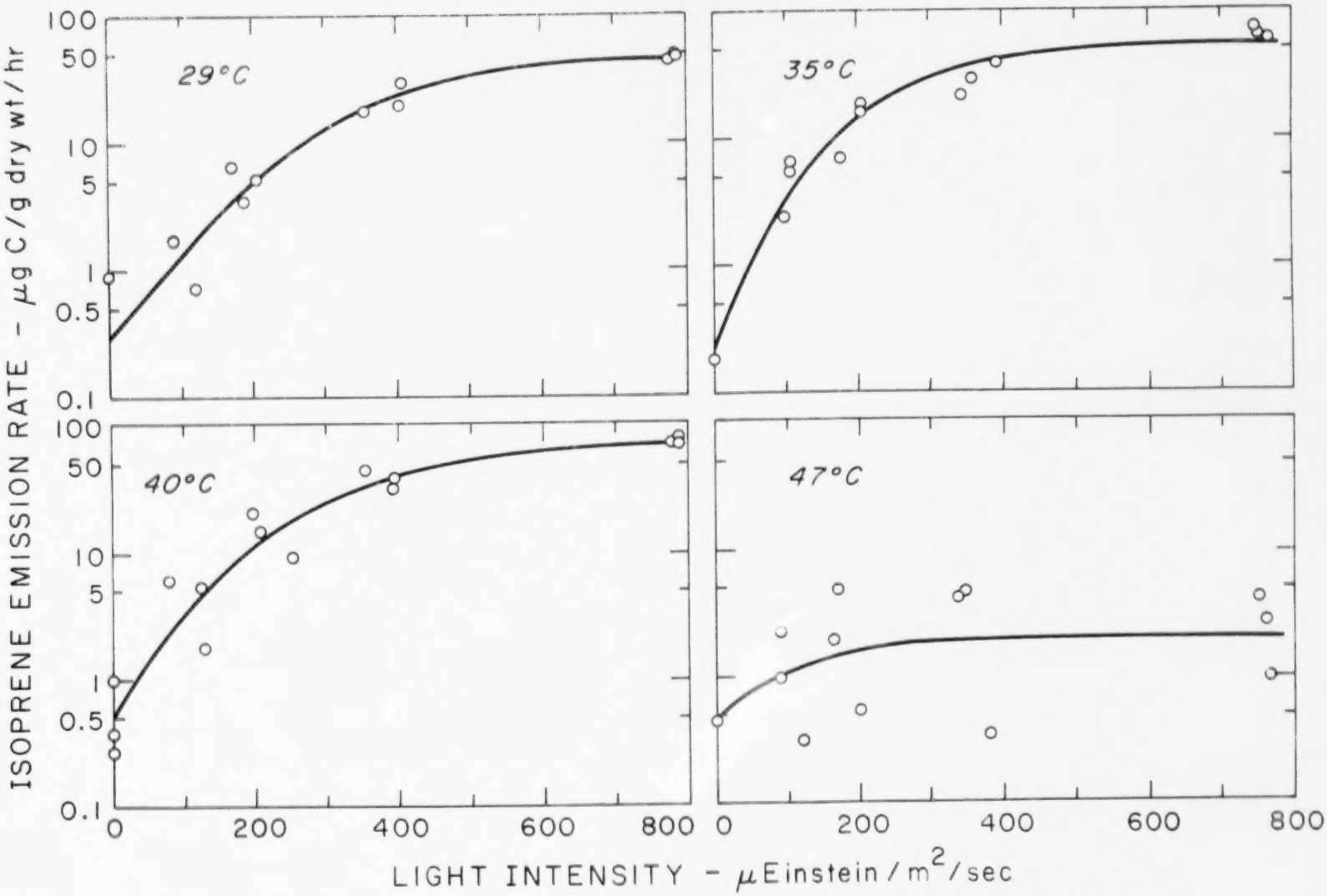

Figure 5. Changes in isoprene emission rates as a function of varying light intensity at 4 leaf temperatures. The data points shown on the graphs are means of triplicate samples.

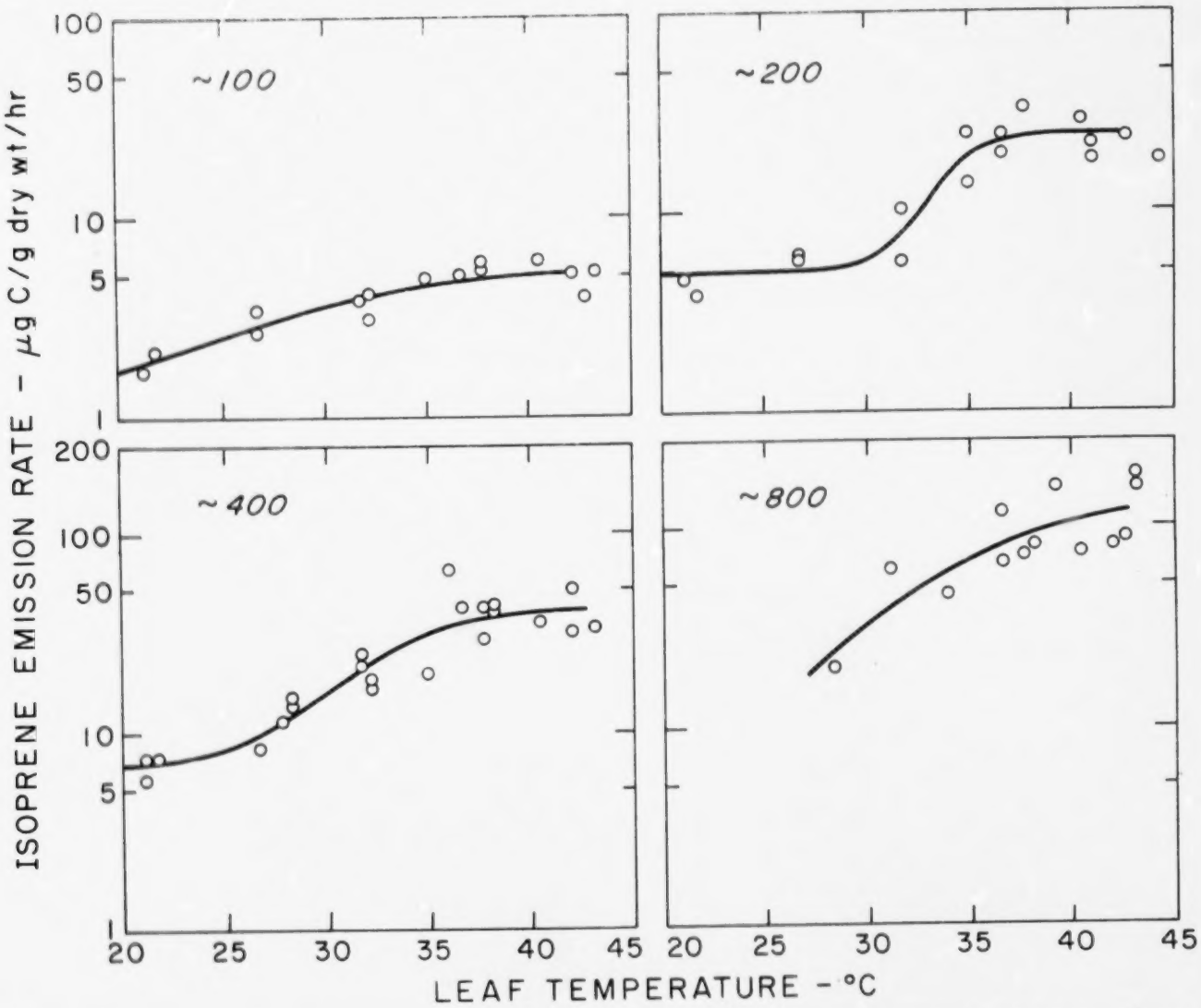

Figure 6. Changes in isoprene emission rates as a function of varying leaf temperature at 4 light intensities. The data points shown on the graphs are means of triplicate samples.

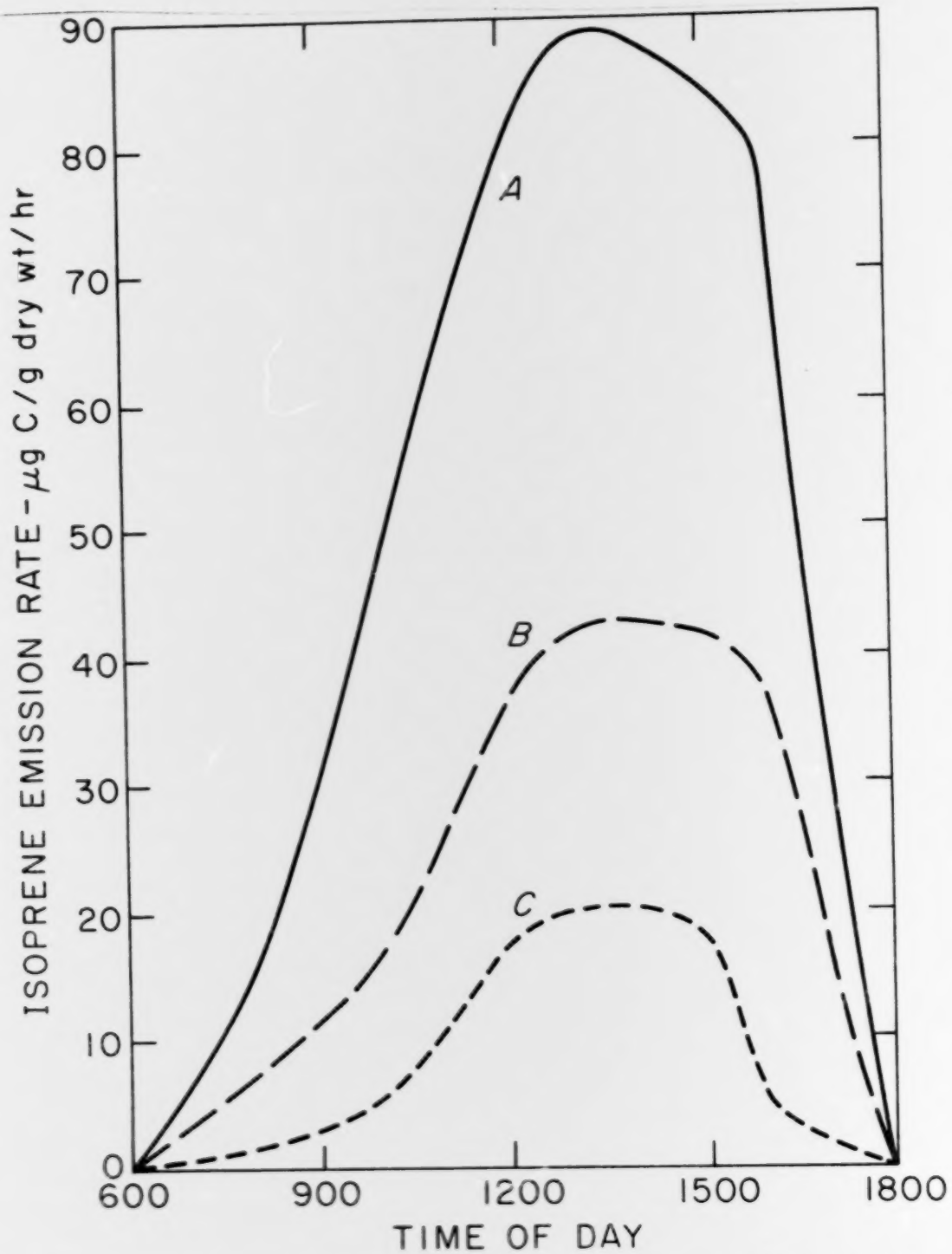

Figure 7. Estimated isoprene emission rates for oak leaves in Tampa Florida for an average of summer days. A--sunlit leaves (assumed leaves hotter than air temperature); B--shaded leaves (assumed leaves 1°C less than air temperature and radiation intensity equal to one-half ambient sunlight); C--shaded leaves (assumed leaves 1°C less than air temperature and radiation intensity equal to 1/4 ambient sunlight). 800 $\mu E/m^2/sec$ equals approximately 0.5 Langley/min.